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Low velocity impact damage in composite laminates based on waste polyolefins

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Abstract

Composite laminates based on polyolefin wastes coming from two different sources are compared with similar systems involving a virgin polypropylene in terms of damage behavior. In particular, this paper reports the response of film-stacked composite laminate plates subjected to falling weight impact tests. As expected, samples based on recycled polyolefins show lower impact parameters with respect to the latter ones even if a partial improvement of the performances can be obtained by modification of the matrix by an adequate coupling agent. Despite the inferior impact behavior, potential uses of new systems provide an opportunity to develop valuable products by reducing the environmental impact of plastic wastes.

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Keywords: Polymer composites; Laminates; Low-velocity impact; Damage; Plastic wastes.

1. Introduction

In the last decades, a growing interest has been dedicated in the use of composite materials for structural applications. In particular, polymer-matrix composites are gaining a special attention to re-place traditional materials in several fields as aeronautical, automotive and marine ones although it is well known that these systems are highly susceptible to internal damage caused by transverse loads even under low-velocity ones. In general, polymer composites can be damaged on the surface but also beneath the surface by relatively light impacts causing barely visible impact damage. Therefore, a lot of studies have been carried out both to highlight effects of several variables linked to constituent properties, geometrical parameters of composite plates and impactor and operative conditions [1-6] as well as to improve the impact response of composite systems and structures [7,8].

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With special regard to thermoplastic based composite items, recent environmental concerns are influencing scientific interest also toward the employment of recycled polymer streams.

In this cases, typical inhomogeneity and incompatibility among different polymers constituting the plastic wastes, especially if coming from municipal sources, and economical constrains usually limit the potential application of discarded plastics. However, these drawbacks are satisfactorily overcome simplifying reprocessing steps and transforming low-cost and abundantly available wastes into valuable new composite materials by use of cheap glass reinforcements [9] but, increasingly, also by natural fibres or fabrics [10-13].

Among available plastic wastes, special attention is paid on the polyolefin fractions mainly obtainable from agricultural field (mulch and greenhouse films), packaging wastes and end-of-life cars.

Taking into account that, due to the interlacing of fiber tows in two directions, woven fabric composites offer better impact resistance as compared to unidirectional composites [14]

The main goal of this work is to present and discuss some experimental results obtained by low-velocity impact testing of polyolefin based laminate plates reinforced with a woven glass fabric in order to highlight potentiality of recycled matrices as valuable alternatives with respect to virgin ones.

All systems were characterized by low-velocity impact tests and the damage behaviour was explored by photographic inspections and morphological analysis.

1. Experimental

2.1 Materials

Investigated systems involve three polyolefin based matrices: a virgin polypropylene PP MA712 (Unipetrol), MFI=12 g/10 min, here coded as *"vPP"*; a regenerated polypropylene (Omega s.r.l.) filled with talc (15-20 wt%), MFI=8 g/10min, coming from bumper scrapes and here indicated as *"R1"* and polyolefin flakes of ~1 cm^2 size from selected post consumer plastic wastes and essentially constituted by a mixing of polypropylene and polyethylene fractions, supplied by Erreplast s.r.l. with a MFI of 5 g/10 min and here coded as *"R2"*.

A commercial polypropylene grafted with 1 wt % of maleic anhydride (PP-g-MA) and commercialized under the trade name Polybond 3200 by Chemtura (Philadelphia) (MFI@190°C, 2.16 kg: 115 g/10 min; density @ 23 °C: 0.91 g/cm^3; melting point: 157 °C) was considered as a compatibilizer to improve the adhesion at the reinforcement-matrix interface for vPP and R1 based composite systems.

Systems compatibilized with 2 wt% of Polybond are identified by the suffix "-PB". Preliminary analysis, not shown in this paper and reported in [15], have shown that the addition of such a coupling agent does not alter the crystallinity of the reference polypropylene phase. At this regard, with the aim to preserve economic aspects of products, given the relatively high cost of analogous commercial coupling agents for polyethylene based systems, these latter ones have been analyzed by now without an adequate compatibilizing additives.

Finally, a plain weave type woven glass fabric (E-type) has been used as reinforcement.

2.2 Fabrication of composite plates

Composite laminates were prepared by alternating film-blown layers of neat or compatibilized polyolefin and glass fibre fabrics according to the traditional film-stacking process and applying a pre-optimized cycle of pressure and temperature as depicted in Figure 1.

The composite laminates, prepared with this procedure and identified in the following by the prefix *"C"*, have an average thickness of 1.3 mm and consist of 8 balanced fabric layers 0°/90°, symmetrically arranged with respect to the middle plane of the laminate ($[(0/90)_4]_s$ configuration).

In all cases the content of reinforcement has been estimated equal to 50 vol% (ASTM D 3171-04, Test Method II) while the void content was less than 1%.

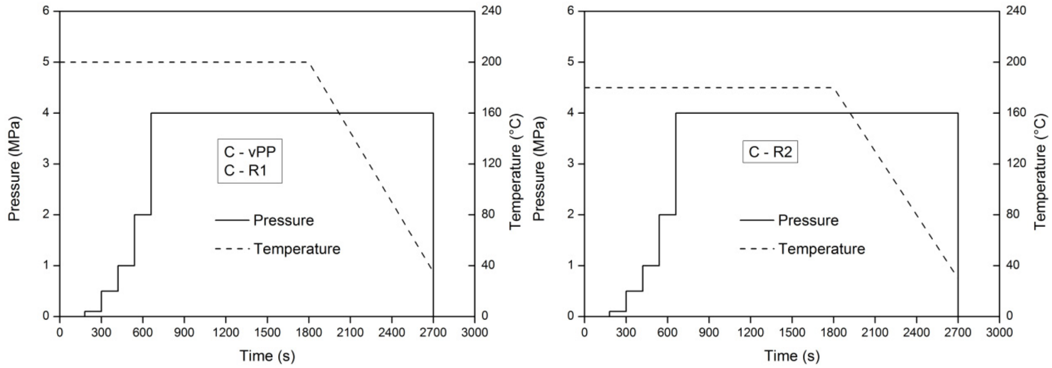

Fig. 1. Operative conditions of the compression molding stage.

2.3 Impact testing

Low-velocity impact tests were performed by using an instrumented free-fall drop dart testing machine (CEAST, mod. “MK16”) that allows to acquire the dart load and the dart velocity history of composite samples. In particular, measurements were carried out according to the ASTM 3029, using an impactor with a hemispherical head having a radius of 19.8 mm and a total mass of 7.64 kg to obtain an impact energy of 30.92 J. Squared samples 130x130 mm^2 were positioned on a ring formed steel support having internal and external diameters equal to 50 mm and 70 mm, respectively. Load-deflection (*L-d*) and load and absorbed energy-time curves were recorded during each test.

Three specimens were tested for each type of sample and the average value of each key impact parameter was reported.

2.3 Morphological observations

The morphology of selected impact section of composite laminates was investigated by using a field-emission scanning electron microscope (SEM) mod. FEI QUANTA 200F.

Samples were sputtered with gold/palladium prior to SEM observations carried out at high vacuum condition and with a voltage of 20 kV.

3. Results and discussion

Figure 2 compares investigated systems in terms of typical load-deflection (*L-d*) curves. The impact energy is such that no rebounding is verified and open curves are always obtained. In particular, considering that analyzed plates are thinner than the radius of the impactor nose, the observed shape of L-d curves is a sign of the occurrence of perforation phenomena the extent of which depends, among others, on properties intrinsically characterizing both matrix and interface. In fact, focusing on the ending part of the descending section of L-d curves, it can be noted that, while for vPP based systems just an initiation of perforation is verified, for other systems the detected trend is typical of frictional effects between the impactor and the specimen.

Alternately, impact data collected so far have been represented as load and absorbed energy as a function of time in Figure 3. Analyzing the picture or, better, evaluating some key-parameters collected in the Table 1, it is clear that the inclusion of compatibilized polypropylene layers reduces the peak load and the time to reach the same with effects higher for vPP based systems with respect to R1 based ones. This behavior can be explained assuming that a worse interfacial adhesion may increase the amount of fibers’ damage and favor pull-out and debonding phenomena responsible for energy absorptions.

Moreover the presence of a significant content of fillers as talc and carbon black, even if well distributed in the recycled polypropylene from car bumpers, may affect the impact resistance of products favoring the crack propagation inside the matrix.

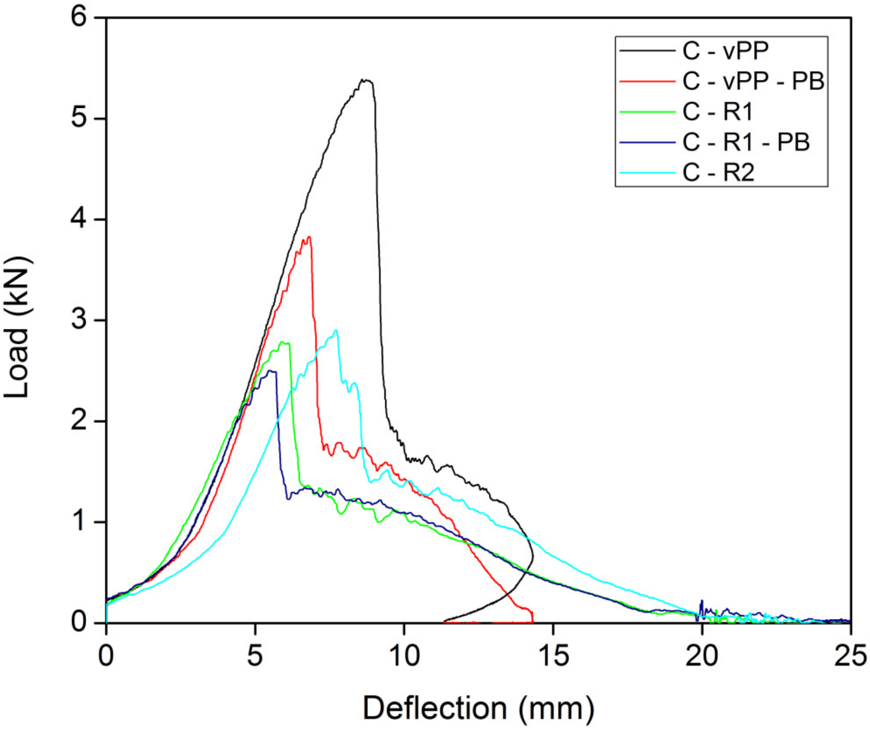

Fig. 2. Load-deflection curves of investigated composite laminates.

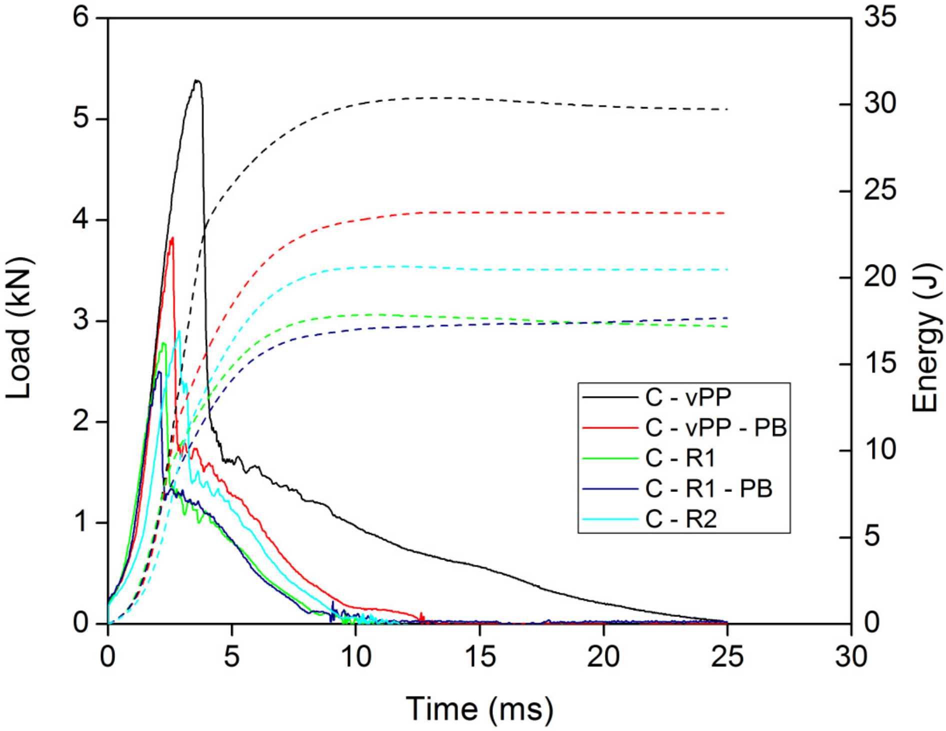

Fig. 3. Load and energy versus time response of all investigated composite laminates.

The low initial slope of the load curve of C-R2 systems can be attributed to an early occurrence of debonding and delaminations, presumably due to the inherent composition of the considered plastic waste (R2), including a high fraction of low density polyethylene, in addition to an expected lower compatibility of the same with the glass reinforcement.

Compatibilizer effects at the interface of polypropylene based systems and mentioned damage mechanisms were confirmed by morphological SEM micrographs reported in Figure 4 a-e. The improved adhesion of matrix on fibers is evident in Figure 4b and 4d relating to systems containing vPP and R1 layers, pre-modified by inclusion of the PP-g-MA compatibilizer, respectively. In not compatibilized laminates, the occurrence of dissipative multiple shear mode of damage is confirmed by evidences of fiber slippages and misalignments especially observed for the composite plates involving layers of as received recycled matrices (see Figures 4c and 4e).

Table 1. Average values of impact parameters characterizing all investigated composite laminates

	Peak Load (kN)	Time to Peak Load (ms)	Deflection at Peak Load (mm)	Energy to Peak Load (J)	Absorbed Energy (J)
C - vPP	5.39±0.12	3.52±0.17	8.57±0.44	19.81±0.75	28.39±1.65
C - vPP - PB	3.79±0.06	2.51±0.14	6.61±0.31	9.64±0.43	23.77±0.72
C - R1	2.79±0.09	2.21±0.14	5.87±0.63	7.54±0.48	17.85±1.13
C - R1 - PB	2.51±0.08	2.03±0.15	5.45±0.41	5.99±0.32	17.72±0.95
C - R2	2.91±0.10	2.87±0.22	7.69±0.75	9.25±0.63	20.65±1.32

Finally, Figure 5 illustrates the top and the bottom surfaces of the investigated laminates. The picture confirms that, at the considered impact energy value, perforation is always verified with effects more marked for not compatibilized composite systems with respect to compatibilized ones. In not compatibilized systems a hemispherical visual damage was observed that means that all the material in contact with the penetrator was permanently deformed. The rhomboidal shape of the damage on compatibilized composite laminates is due, on the contrary, to a recover of the deformation in the area not interested by the fibre failure. In the latter case, by observing the C-vPP and C-R2 specimens against the light, a very limited extension of the delamination was noted. However, this is a very interesting point that need deeper analysis.

The size of cracks along the two directions is comparable with the impactor diameter denoting the low tendency of these materials to delamination. This assertion was confirmed by visual analysis: a light source against the C-vPP, C-vPP-PB and C-R2 specimens, on the opposite side respect to the observation one, evidenced no delaminated area outside the impacted one. The same was not possible to do on the C-R1 and C-R1-PB laminates since their non trasparency.

4. Conclusions

The behavior of thermoplastic composite laminate plates subjected to low velocity impact loading has been examined to highlight potentialities of recycled matrices with respect to virgin ones. Attention was focused on polyolefin based composite systems reinforced by a woven glass fabric and involving, as the matrix, two different type of recycled polyolefin streams: end-of-life car bumpers (R1) and bottle wastes (R2). Composite plates obtained by a typical film stacking route were compared in terms of low velocity impact properties with similar items based on neat virgin polypropylene (vPP). Moreover, the effect of a coupling agent as a modifier of plastic layers in case of vPP and R1 matrices was also taken into account.

Impact results, confirmed by SEM morphological observations, showed that the improvement of the interfacial adhesion prevent not only the fiber breakage but also the occurrence of typical dissipative mechanisms of damage under the applied testing conditions.

Minor impact resistance was shown by composite laminates including plastic layers from bottle wastes given the intrinsic formulation of the matrix mainly constituted by low density polyethylene fractions and, do far, the lack of consideration of an appropriate compatiblizer.

Finally, visual inspection of impacted areas have highlighted the low tendency of investigated systems to delamination as well as the occurrence of permanent deformations of the material in contact with the penetrator, partially recovered in compatibilized laminates.

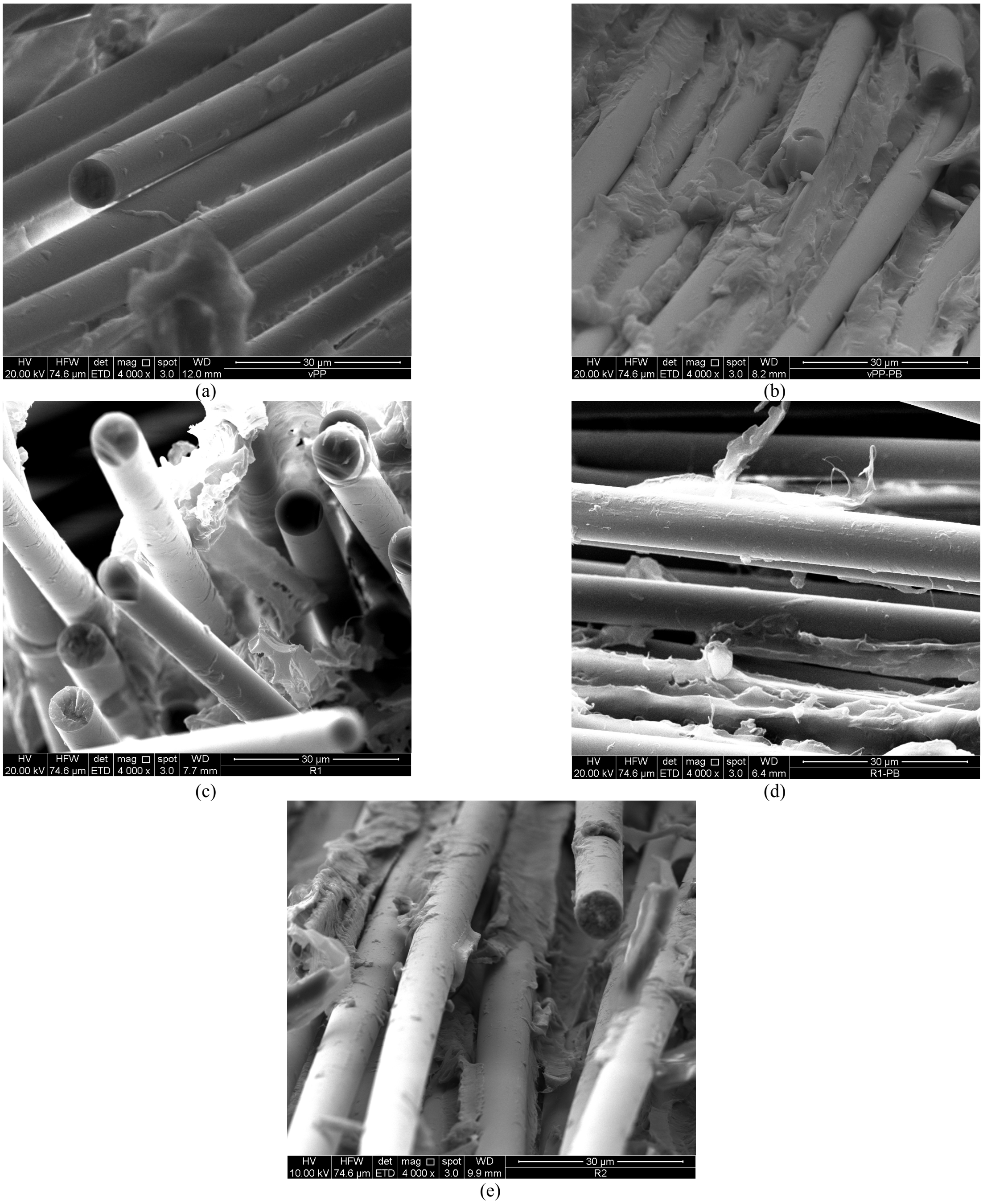

Fig. 4. SEM micrographs of impacted areas of (a) C-vPP, (b) C-vPP-PB, (c) C-R1, (d) C-R1-PB, (e) C-R2 systems.

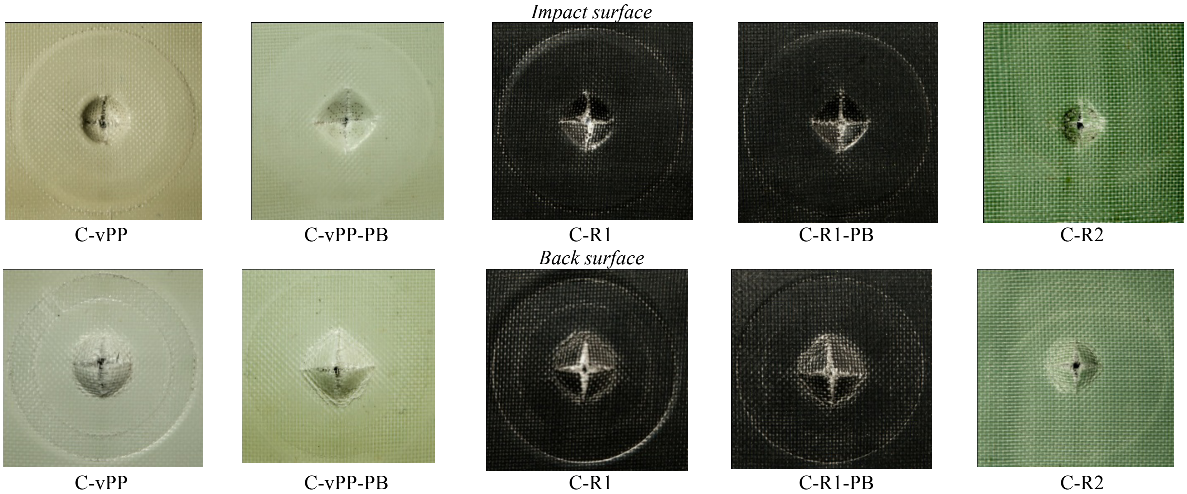

Fig. 5. Photographic images of top and bottom surfaces of impacted composite plates.

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